

# High Temperature Carbonization (HTC)

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**Partner:**

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ORNL is managed by UT-Battelle LLC for the US Department of Energy

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# Overview

## Timeline

- Project Start: 11/17/20\*
- Project End: 9/30/22
- Progress: 66%

\* Due to internal delays

## Budget

Overall budget:

- FY21 – FY22: \$3.5M

### Detail:

- FY21: \$2.0M
- FY22: \$1.5M

## Barriers

- Barriers addressed
  - Cost: A goal of this project is to reduce energy consumption in the carbon fiber conversion process and therefore total carbon fiber cost.
  - Inadequate supply base: Another goal of this project is to reduce the required processing time for carbonization and therefore increase overall throughput.

*2017 U.S. DRIVE MTT Roadmap Report, section 4*

## Partners

- Project lead: Oak Ridge National Laboratory (ORNL)
- Partner: 4XTechnologies, LLC  
(formerly RMX Technologies)

# Relevance

- Objectives:

Lower the cost of production of Carbon Fiber (CF) with a novel method for High Temperature Carbonization (HTC):

- Based on **dielectric heating**.
- **Allowing faster** and more efficient conversion than conventional process.
- Operating at **atmospheric pressure**.
- Scale the technology to a nameplate capacity up to one annual metric ton by project end date.

- Impacts:

- Reduce unit energy consumption of HTC stage (kWh/kg) by ca. 20% (ca. 5% of the cost reduction on the carbon fiber (CF) overall manufacturing process).
- Produce equal or better quality carbon fiber.

# Milestones FY2022

All **FY2021** milestones (MS M1, M2, M3 and M4) were completed before Dec. 31, 2021

Date	Milestone FY2022	Status
Feb. 28, 2022	<b>M5:</b> Process 4 tows of 24k filaments ea. CF production (Density = 1.7 g/cc, tensile strength = 400ksi, modulus = 25Msi, residence time = 200s)	Completed
Apr. 30, 2022	<b>M6:</b> Process 4 tows of 24k filaments ea. CF production (Density = 1.7 g/cc, tensile strength = 500ksi, modulus = 27Msi, residence time = 200s)	Completed
June 30, 2022	<b>M7:</b> Process 4 tows of 24k filaments ea. CF production (Density = 1.7 g/cc, tensile strength = 550ksi, modulus = 29Msi, residence time = 160s)	Partially completed, In progress
Aug. 31, 2022	<b>M8:</b> Process optimization. CF production (tensile strength = 550ksi, modulus = 29Msi). Energy balance acquisition.	N/A
Aug. 31, 2022	<b>M9:</b> Demonstrate HTC at least 5% cost savings versus conventional carbonization.	N/A
Sept. 30, 2022	<b>M10:</b> Final report with discussion about further technology implementation/scale up.	N/A

NOTE: The HTC project experienced two significant delays:

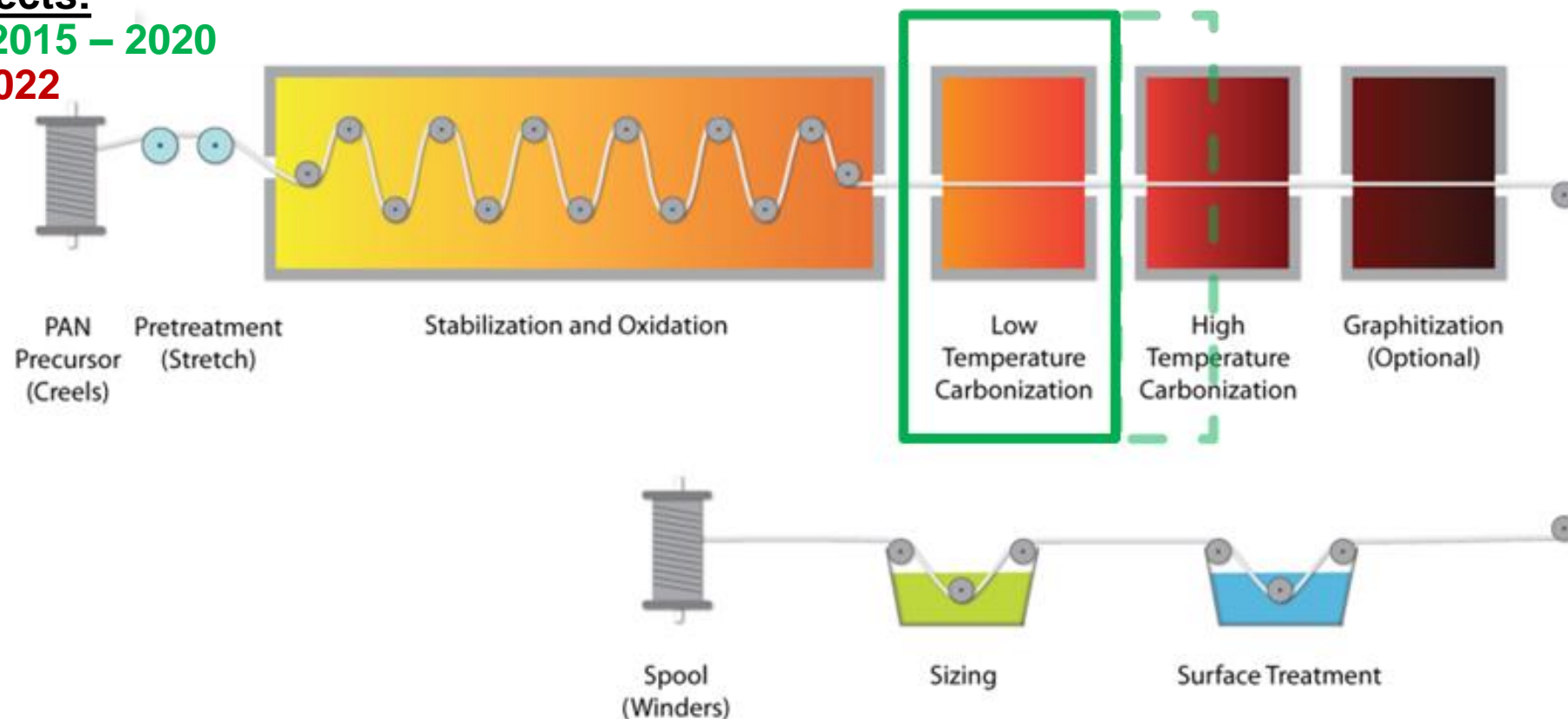
- 7 weeks at the beginning (contract between ORNL and 4XTechnologies delayed)
- 5 weeks in July 2022 (machined parts for EM applicator).

# Approach (conventional PAN processing)

Two VTO projects:

LTC (CPEC): 2015 – 2020

HTC: 2021 - 2022



## Major Manufacturing Costs

Precursor	ca. 46%
Oxidative stabilization	18%
Carbonization	24%
Other	12%

- Automotive cost target is \$5 - \$7/lb
- Tensile property requirements (DOE general guideline) are 250 ksi, 25 Msi, 1% ultimate strain
- ORNL is developing major technological breakthroughs for major cost elements

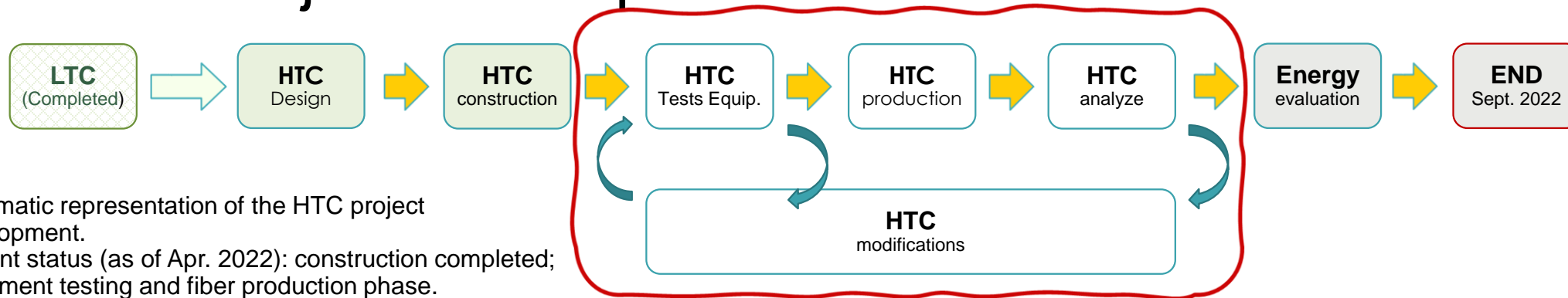
# Approach

- Conventional furnaces consume significant energy heating large volumes of inert gas surrounding the fiber.
- If thermal energy could be directly coupled from an energy source to the fiber, tremendous energy savings could be realized.
- This project uses electromagnetic coupling to directly and indirectly heat the fiber.
- Dielectric/Maxwell-Wagner heating mechanisms are utilized

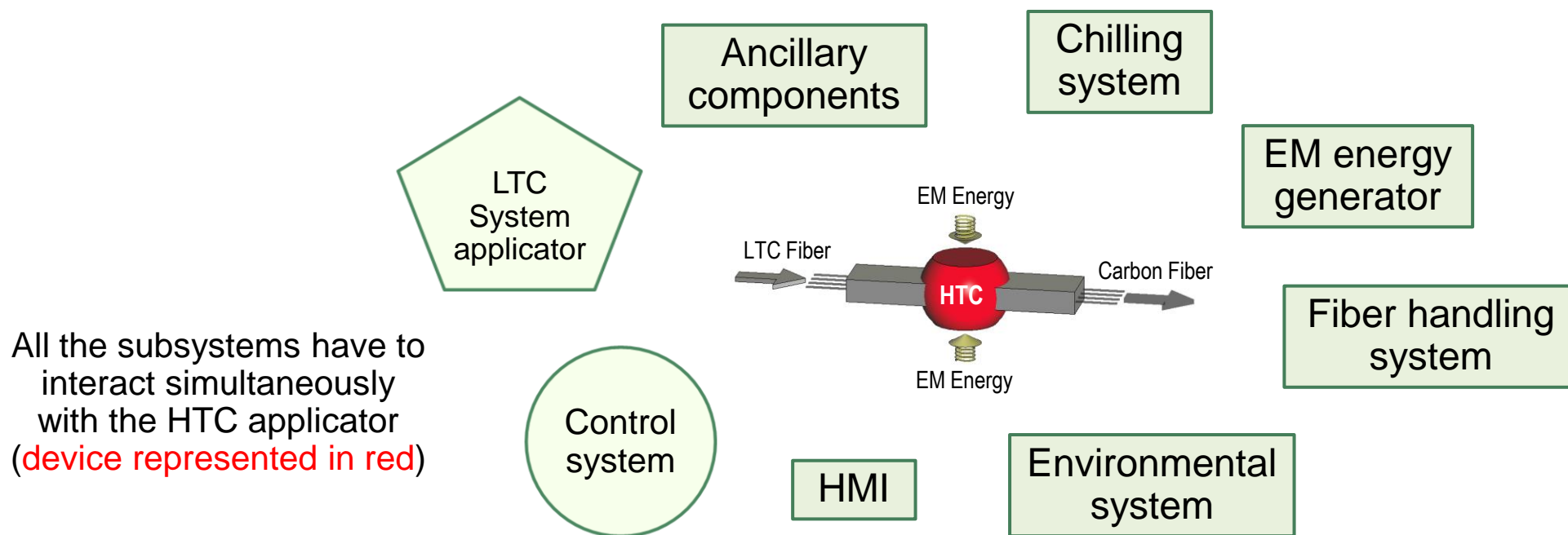
$$P_v = 2\pi f |E|^2 \epsilon_0 \epsilon' \tan \delta$$

- $P_v$  volumetric power transferred to the material.
- $\epsilon'$  is the relative dielectric constant.
- $\epsilon_0$  is permittivity of free space,  $8.85418782 \times 10^{-12}$  F/m.
- $|E|$  is the magnitude of the local electric field intensity (V/m).
- $\tan \delta$  is the loss tangent of the material.
- $f$  is the operational frequency.

# Progress — Project development



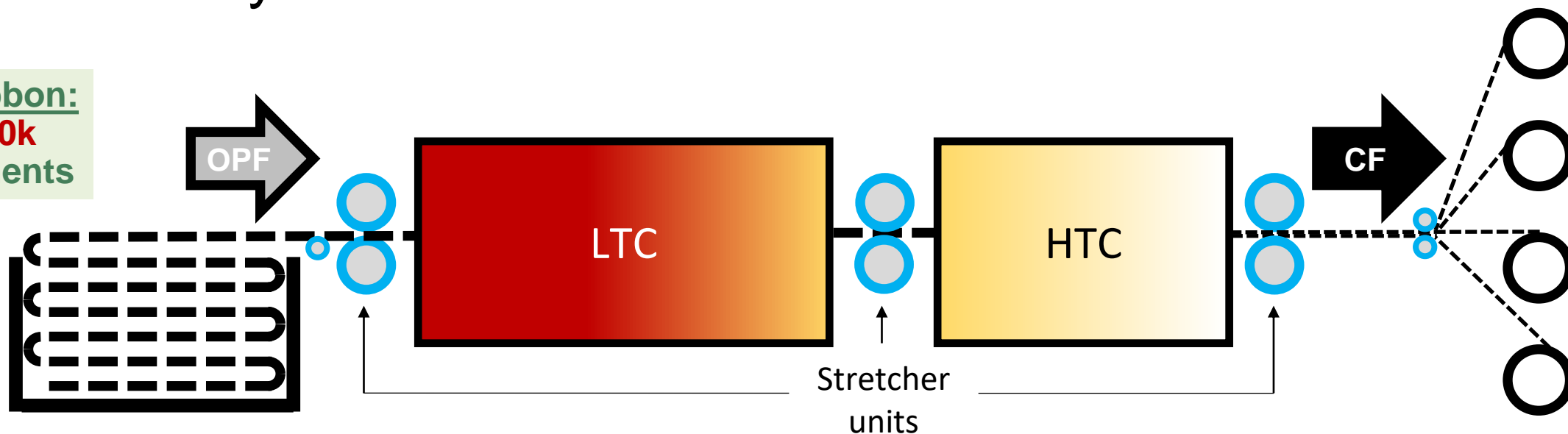
- Representation of the applicator and all its subsystem:



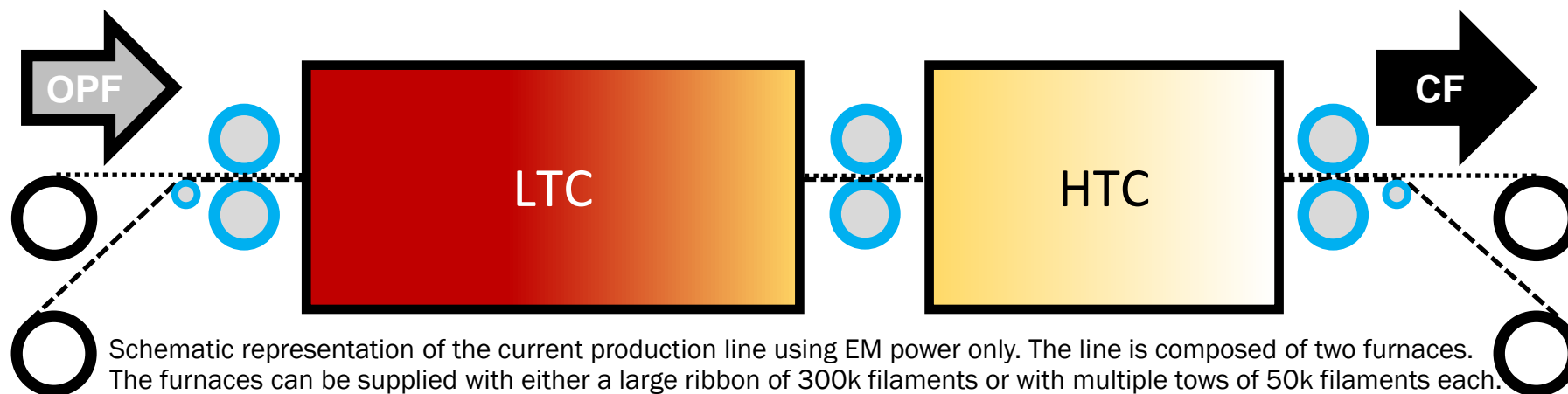


# Progress — Layout

1 Ribbon:  
**300k**  
filaments



2 x 50k  
Tows:  
**100k**  
filaments  
total



Schematic representation of the current production line using EM power only. The line is composed of two furnaces. The furnaces can be supplied with either a large ribbon of 300k filaments or with multiple tows of 50k filaments each.  
Upper representation: usage of **1 large ribbon**.  
Lower representation: usage of **two tows**.



# Progress — Results

## Mechanical properties (single filament testing):

### 1 Ribbon: **300k** filaments

Four sampling locations on the 300k filament ribbon				
Sampling location	Diameter [μm]	Tensile Strength [ksi]	Modulus [Msi]	Strain [%]
Off center	8.3	380	29.3	1.25
	0.4	156	1.2	0.45
Central zone (damaged area)	8.5	334	27.7	1.2
	0.2	123	0.9	0.4
Right side	8	327	29.5	1.08
	3.2	104	1.3	0.27
Left side	8.3	269	27.9	0.96
	0.3	134	0.9	0.44

This results met milestones M4 (Dec. 2021) requirements;

### **M4 (Go/No-Go):**

CF production (1.6g/cc, 300 ksi tensile strength, and 25 Msi modulus).

### 2 Tows: **100k** filaments

Tow	Fiber type	Sample [#]	Diameter [μm]	Tensile strength [ksi]	Modulus [Msi]	Strain at breakage [%]
#1	LTC	1	8.32 0.2	198.44 39.18	8.07 0.22	2.46 0.47
	HTC	2	6.84 0.36	536.98 124.27	33.05 1.32	1.57 0.33
	HTC	3	6.99 0.46	514.92 146.12	31.05 1.21	1.6 0.43
	HTC	4	6.48 0.61	595.57 88.31	30.75 1.21	1.87 0.25
	HTC	5	6.92 0.24	514.88 152.9	29.32 1.26	1.68 0.44
#2	LTC	1	8.28 0.4	187.89 54.99	9.37 1.76	2.07 0.71
	HTC	2	6.8 0.49	512.48 166.19	32.15 0.87	1.55 0.49
	HTC	3	6.8 0.52	568.89 105.15	31.48 0.67	1.75 0.32
	HTC	4	6.58 0.46	559.88 107.2	31.06 1.35	1.72 0.29
	HTC	5	7.59 0.29	427.17 139.5	23.74 2.17	1.73 0.5

Residence time defined by colors: Yellow > Red > Brown > Green.  
POWER input: was kept CONSTANT for ALL samples.

**Green and Brown:** total carbonization time (LTC + HTC) = **less than 1 minute**.

Note: During this process, we observed inconsistency in the positioning of the fiber inside of the applicator. This shortcoming will be corrected in further experimental work.

# Progress — Results vs. Milestones

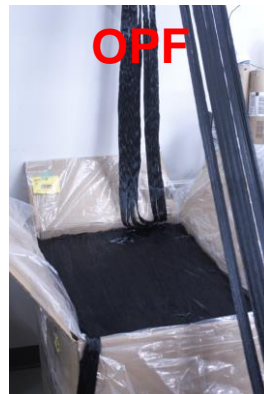
Date	Milestone FY2022	Status	Based on data from prior viewgraph
Feb. 28, 2022	<b>M5:</b> Process 4 tows of 24k filaments ea. CF production (Density = 1.7 g/cc, tensile strength = 400ksi, modulus = 25Msi, residence time = 200s)	<b>Completed</b>	<i>Based on data from prior viewgraph is completed</i>
Apr. 30, 2022	<b>M6:</b> Process 4 tows of 24k filaments ea. CF production (Density = 1.7 g/cc, tensile strength = 500ksi, modulus = 27Msi, residence time = 200s)	<b>Completed</b>	<b>Density:</b> all samples > 1.7 (not reported in table) <b>Res. Time:</b> Red, Brown, Green < 200s <b>Strength:</b> Yellow, Red, Brown > 500 ksi <b>Modulus:</b> Yellow, Red, Brown > 27 Msi → MS 6 completed
June 30, 2022	<b>M7:</b> Process 4 tows of 24k filaments ea. CF production (Density = 1.7 g/cc, tensile strength = 550ksi, modulus = 29Msi, residence time = 160s)	<b>Partially completed</b> <ul style="list-style-type: none"> <li>• The Brown samples meet all Milestone M7 requirements.</li> <li>• Process shows larger deviation at short residence time (Green samples).</li> <li>• In progress / further evaluation needed, challenges to be addressed, next Vg).</li> </ul>	<b>Density &gt; 1.7</b> (not reported in table) <b>Res. Time:</b> Red, Brown, Green < 160s <b>Strength:</b> Red (tow #2) Brown (tow #1 and #2) > 550 ksi <b>Modulus:</b> All samples except Green (tow #2) > 29 Msi → MS 7 partially completed

# Progress – Results and remaining challenges

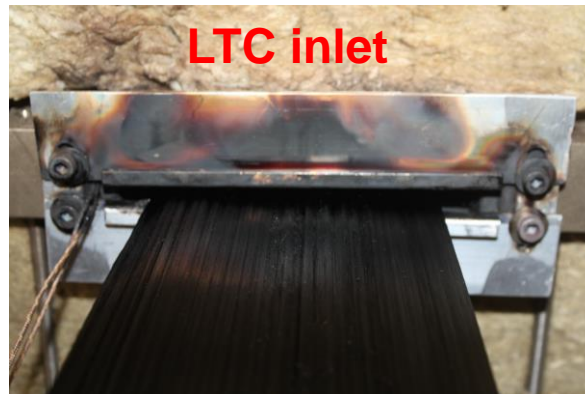
- Processing fiber continuously requires overcoming typical challenges

- 300k ribbon issues

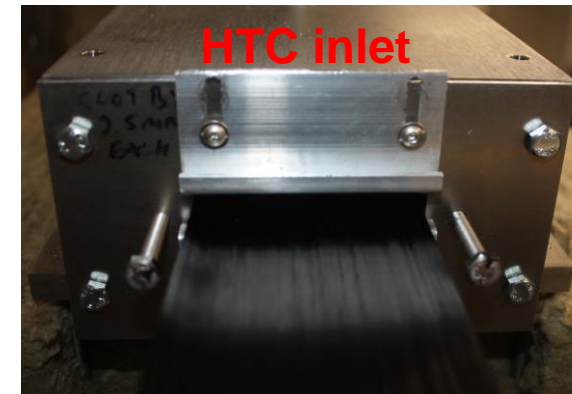
- Proper fiber insertion.
- Proper alignment.



OPF feedstock  
(300k filament  
ribbon).



Inlet of **LTC furnace**. The presentation is acceptable. This part of the process is well known and smooth.



Inlet of **HTC furnace** (narrower than LTC). The setup was **designed for 4 x 50k filaments**: with a 300k ribbon, it is **overloaded**.

- Proper process.
- Proper tow separation.
- Proper fiber take-up.



Damaged might happen during HTC...



... making the split difficult...



... and the take up impossible.

# Progress – Results and remaining challenges (cont.)

- Cavity tuning performance optimization

Experiments indicate:

- The cavity works as is and can produce fiber exceeding the criteria of programmatic milestones to date. However:
- Additional experimental test must be conducted to get a better knowledge of the cavity behavior (performance, tuning capability, temperature distribution, etc.).
- Depending upon evaluation, some modification may be implemented:
  - CEM modeling will be needed:
    - Part of the complexity might not have been taken into account during former modeling activity (energy delivery and distribution to/in the applicator, and geometry of the applicator itself).
  - Potentially, better system performance might be achieved.

# Response to Previous Year Reviewer's Comments

- Previous year scoring (AMR 2021):

- 100% of reviewers indicated that the project was relevant to current DOE objectives.
- 75% of reviewers indicated that the resources were sufficient for the challenge tasks, and only 25% of the reviewers indicated that the resources maybe excessive.
- Researcher answers to reviewer in “green”.

- Question 1:

*Approach to performing the work—the degree to which technical barriers are addressed, the project is well-designed, and well planned.*

- R #1, R #2 and R#3: Favorable comments Thank you.
- R #4: “The scientific approach seemed reasonable to the reviewer... How that impacts energy was not clear to this reviewer” Compared to conventional process, the energy consumption can be reduced by lowering the amount of energy per mass unit of produced material (direct energy coupling, better energy confinement, shorter residence time, reduced maintenance, shorter down time).

- Question 2:

*Technical Accomplishments and Progress toward overall project goals—the degree to which progress has been made and plan is on schedule.*

- R #1 and #3: Favorable comments Thank you.
- R #2: “Progress of the project is good... [but] wondered whether the PIs will be able to meet [the] deadline due to the time lost to COVID-19.” Thank you. As of Apr. 2022, most of the MS were met.
- R #4: “... Also, the team proposes to use Composite Epoxy Material (CEM) to establish the design methodology (by the way, CEM was never explained in slides).” True: CEM was not explained in the slides, and we apologize for this. CEM stands for Computational ElectroMagnetics. This acronym could have been misleading.



# Response to Previous Year Reviewer's Comments

- Question 3:

*Collaboration and Coordination Across Project Team.*

- R #1 and #2: Favorable comments Thank you.
- R #3: *"The reviewer identified synergy between ORNL and 4XTehnologies. the relative amount of work being carried out by each organization was not explained in more detail."*
- R #4: *"The presenter needs to better communicate the role and importance of 4XTechnologies."*

Answer reviewers 3 and 4:

- The collaboration between ORNL and 4XTechnologies is ideal for this type of project: ORNL brings an extensive background and knowledge in material science/CF and their evaluations. 4XT brings engineering background in plasma physics, electromagnetics (including modeling), and hardware construction.

- Question 4:

*Proposed Future Research—the degree to which the project has effectively planned its future work in a logical manner by incorporating appropriate decision points, considering barriers to the realization of the proposed technology and, when sensible, mitigating risk by providing alternate development pathways.*

- R #1 and #2: Favorable comments Thank you.
- R #3: "... Perhaps that decision point [the Sept. 30 Go/No-go milestone] needs to be postponed." Because the projects experienced an additional delay of more than a month in the July-August period, milestone M4 (Go/No-go, Sept. 30) had to be postponed to Dec. 31, 2021.
- R #4: "... there are no clear decision points to measure progress of the design and build to make sure it is on track to meet milestones." FY2021 was mostly dedicated to design and construction. There was no way to measure the scientific progress of the project (based on material properties) without the equipment being built. Progress could only be evaluated based on task completion. However, FY2022 being dedicated to experimental work, can be evaluated based on scientific and technical metrics.

# Response to Previous Year Reviewer's Comments

- Question 5:

*Relevance—Does this project support the overall DOE objectives?*

- R #1: “Lowering the amount of energy used in CF production [...] aligns directly with DOE goals.”
- R #2: “The cost of CF is one of the barriers to adoption in high-volume [...] reducing the material cost by 5% is a required improvement...” In agreement.
- R #3: “... CF cost is a major barrier in wider adoption in vehicles...” In agreement.
- R #4: “Seems very small to the reviewer for the cost and level of effort” This can be answered more exactly with an economical evaluation of the overall carbonization process.

- Question 6:

*Resources—How sufficient are the resources for the project to achieve the stated milestones in a timely fashion?*

- R #1: “... pretty high for the return on investment [...] regarding a] speculation of 5% energy savings”. High cost of this project is due to the use of new technology, EM modeling, custom equipment (power supplies, construction of an applicator, control system, safety, lab space modification, etc.).
- R #2: “... have necessary resources” Agree.
- R #3: “The remaining funds [...] appeared to be sufficient for project plan and challenges.” Agree.
- R #4: “It is difficult to tell at this point if the funds are sufficient to carry out this project. The ratio of work done to money spent has not been presented.” The first year was dedicated to modeling, design, equipment selection and procurement, and construction. The investment matched with project progress.



# Collaboration and Coordination

## Collaboration detail:



- Extensive background in material science.
- Solid expertise in carbon fiber R&D, manufacturing.
- Extensive capability for material testing and evaluation.
- Project management and project guidance.



## Contractor, outside VTO

- Electromagnetism and plasma: CEM work.
- General engineering.
- Hardware construction.
- Experimental work.

*4XTechnologies is a dynamic startup located in Knoxville, TN.*

**Former collaborations between ORNL and 4XTechnologies have demonstrated that this partnership is ideal for this type of project.**

# Remaining Challenges and Barriers

- Based on past experience and the knowledge acquired with the experimental development of HTC project, anticipated barriers could be:
  - Better selection of dielectric materials.
  - Improve temperature monitoring inside the applicator .
  - Further development of CEM modeling that encompasses tuning performances and thermal property changes.
  - Proper selection of materials for components exposed to high temperature ( $>1000^{\circ}\text{C}$ ).

# Proposed Research

## – FY2022: (fiber production)

- Improvement of the fiber movement control during processing.
- Cavity tuning performance evaluation.
- Implement all needed modification towards optimization of the HTC process.
- Energy consumption evaluation (milestones M8 and M9).
- Intent to reach 700ksi strength, 33Msi modulus (not a programmatic milestone, time permitting).
- Final report (M10).

# Summary

- HTC project initiated on Nov 17, 2020.
- Initial stage of design, modeling, construction was completed on Sept. 2021.
- Commissioning and equipment performance evaluation:
  - Oct to early December 2021.
  - First equipment modification(s) were implemented during this time.
- All MS 2021 M1 to M4, including Go/No-Go (300ksi).
- Fiber production and evaluation:
  - Since December 2021.
  - Mid April: 500ksi strength CF were achieved. (validation of M5 and M6)
- HTC project resolved former delays:
  - In spite of contractual and supplier delays, project is *back on original schedule*. (as of Apr. 2022)
- HTC project is on track for success.





**Questions?**

Thank  
you for  
your  
attention

# Technical Backup

# Milestones (FY2021)

Date	Milestone FY2021	Status
Dec. 30, 2020	<b>M1:</b> Data and design Evaluation between two applicator configurations. Down selection of one of them to go to HTC.	Completed: Feb. 26, 2021
March 30, 2021	<b>M2:</b> design with CEM modeling completed. Beginning of procurement.	Completed
June 30, 2021	<b>M3:</b> Commissioning of HTC. Stable operation (15min).	Completed
Sept. 30, 2021	<b>M4 (Go/No-Go):</b> CF production (1.6g/cc, 300 ksi tensile strength, and 25 Msi modulus).	Rescheduled
Dec. 31, 2021	<b>M4 (Go/No-Go):</b> CF production (1.6g/cc, 300 ksi tensile strength, and 25 Msi modulus).	Completed